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Graphene-Based Nanomaterials for Dye Removal from Wastewater: A Review

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ABSTRACT

Among those, the release of synthetic dyes from textile, paper, leather, and cosmetic industries into water bodies poses a serious environmental problem because of their toxicity, non-biodegradability, and carcinogenic nature. In many cases, conventional wastewater treatment technologies have shown limited abilities to remove such a complex organic pollutant; therefore, the pursuit for advanced materials is under way. Graphene-based nanomaterials including graphene oxide, reduced graphene oxide, and their composites have emerged as promising adsorbents and catalysts for dye removal due to their ultra-high surface area, tunable surface chemistry, and robust mechanical strength. Generally, the main dye sequestration mechanisms involve: π-π stacking, electrostatic interaction, and hydrogen bonding. This review will cover in detail dye classification, conventional removal methods, the synthesis and functionalization of GBNs, their application, the dominating removal mechanisms, factors affecting efficiency, adsorption kinetics, and isotherms. Comparative studies between the prepared GBNs and some other adsorbents prove the superior performance of GBNs. Finally, the critical challenges related to scalability, environmental fate, and ecotoxicity are discussed in this review; it also proposes future research directions that could help the practical application of these nanomaterials in sustainable remediation.

INTRODUCTION

The largely uncontrolled discharge of synthetic dyes frequently is one of the most serious problems in the management of industrial wastewater. As complex organic compounds utilized in large quantities particularly by the textile industry to satisfy roughly two-thirds of the 700,000+ tons of synthetic dyes produced annually on a global scale, combined with paper printing, leather processing, food products, and cosmetics, synthetic dyes are intended to display maximum durability in response to periods of light exposure, oxidation, and microbiological degradation (1), (2). A defining feature of synthetic dyes and the core source of the unresolved dilemma is the detrimental influence on the overall well-being of aquatic food chains owing to the capability of synthetic dyes to absorb and scatter irradiation from the sun. More significantly, synthetic dyes contain a large number of toxic aromatic amines, mutagens, and carcinogens that directly endanger the well-being of plant and animal life in water bodies and the public via bioaccumulation (3,4). Long-term exposure is thought to influence the onset and progression of a wide range of disorders from human exposure to synthetic dyes, including human skin irritation, renal toxicity, neuropsychological disturbances, and cancer.

The commonly used wastewater treatment technologies, though considered to be the foundation, have often been inadequate in dye removal. Physical treatments, like adsorption by activated carbon, when implemented, show significant efficiency but possess a high operational cost accompanied by activator regeneration problems (5). Filtration membranes, including nanofiltration and reverse osmosis, though showing significant dye removal efficiency, experience high operational costs accompanied by fouling (6). Chemical treatments, including coagulation-flocculation and AOPs, though displaying significant efficiency, experience high operational costs accompanied by a high volume of toxic sludge formation or the use of expensive chemicals, along with intermediate compound formation that has a greater toxicity level than the original dye (7,8). Biological treatments, including bacteria, fungi, and algae, though showing significant efficiency and eco-friendliness, experience low operational efficiency, long periods, and low efficiency with non-biodegradable dye, as well as inhibitory compounds (9).

The significant advances in nanotechnology have led to a paradigm in environmental remediation: nanomaterials, with superior surface properties and tunable electronic properties, have unlocked unparalleled opportunities for wastewater treatment and pollution control. Among such nanomaterials, graphene-based nanomaterials (GBNs) have taken center stage since the groundbreaking observation of graphene in 2004 (10). Analogous to graphene, GBNs such as graphene oxide (GO) and reduced graphene oxide (rGO) have been found to inherit an ultra-high theoretical surface area of approximately 2630 m^2/g according to their surface chemistry derived from a configuration formed by a single atomic layer of sp^2-hybridized carbon atoms in a two-dimensional structure. GBNs, owing to their superior surface properties and reactivity, can be employed as efficient sorbents themselves and as effective supports for catalyst particles to unlock new avenues in "sorb-and-degrade" strategies (11). The goal of this review is to conduct an in-depth, systematic review of GBNs as an effective means to remove dyes from wastewater. It will discuss the synthesis and effective functionalization strategies for GBNs, reveal the underlying physical and chemical properties to drive effective sorptive action, and finally discuss current environmental implications and challenges in terms of GBNs to fill any existing gaps between recent advances and real-world implications (12).

CLASSIFICATION OF DYES IN WASTEWATER

A deep understanding of the dye chemistry is essential for the effective design of removal strategies, as the physicochemical properties of dyes play a critical role in determining their interactions with the treatment materials. Dyes are classified based on the following criteria:

Chemical Structure

This is the most fundamental classification of dyes. The chemical structure of any dye defines its color, stability, and potential for interaction

- **Azo Dyes (-N=N-):** This largest and most commercially important group ('>60%) is employed on cotton, wool, and leather products. Their persistence in the ecosystem and the production of carcinogenic aromatic amines upon anaerobic degradation (for instance, benzidine) have made them a priority pollutant (13).
- **Anthraquinone Dyes:** The second most important group, valued based on âcustomary bright tintfastnessâ. They have a sophisticated structure and are more resistant to deterioration than azo dyes (14).
- **Triarymethane Dyes:** These dyes are known to have strong colors with low lightfastness. Suitable examples are Malachite Green and Crystal Violet, which are toxic and used in the paper, silk, and leather industries (15).
- **Phthalocyanine and Xanthene family of dyes:** The former are metal complex dyes possessing good stability, while the latter are fluorescent dyes used in paper and cosmetic products (16).

Ionic Nature (Charge in Aqueous Solution)

This property has critical implications for removal by electrostatic interactions.

- **Cationic (Basic) Dyes:** Possess a positive charge on the chromophore. Examples include Methylene Blue and Safranin O. These are visually impactful and toxic at low concentrations (17).
- **Anionic Dyes:** These have a negative charge due to the presence of the sulfonic group ($-SO_3^-$) and the carboxyl group ($-COO^-$). This is a large group that includes Acid dyes used on wool/nylon fabric, Reactive dyes that covalently bind to cellulose, and Direct dyes on paper/cotton (18).
- **Non-ionic (Disperse) Dyes:** Very low solubility in water. Used in synthetic fibers like polyester. These are used as a fine dispersion.

Application and Industrial Source

This practical classification enables dye types to be related to their industrial origin (19).

- **Reactive Dyes:** Mainly used for cotton fabrics, i.e., a major portion of textile effluents. The hydrolyzed form of these dyes that cannot bind to fabrics is the major pollutant.
- **Acid and Basic Dyes:** For the protein fibers (wool and silk) and acrylics, commonly found in tannery and textile wastes (20).
- **Direct and Vat Dyes:** Used in Cotton, Cellulosics, paper industry, Textile Industry etc.
- **Disperse dyes:** One of the major constituents of wastewater from polyester or acetate fibers treatment (21).

CONVENTIONAL DYE REMOVAL TECHNIQUES: AN OVERVIEW

A critical review of existing techniques thus has intrinsic disadvantages that are the driving force behind the formulation of improved nanomaterials, such as GBNs.

Physical Treatment Methods

- **Adsorption:** Although the benchmark adsorbent is activated carbon, because of its porosity, the non-selective nature, associated high cost, and the important capacity loss during thermal regeneration limit its economic feasibility to large-scale dye removal (22).

- **Membrane Filtration:** Membrane processes such as ultra-filtration (UF), nano-filtration (NF), and Reverse Osmosis (RO) are capable of producing high-quality effluent. However, they are still suffering from membrane fouling problems, high capital and operation energy costs, as well as producing concentrated brine streams that require further processing (23).
- **Coagulation-Flocculation:** Inorganic coagulants (alum, ferric chloride), or organic polymers, are used to destabilize and aggregate dye particles for sedimentation. This method is effective only for some dyes, producing voluminous, hazardous sludge, and is inefficient for soluble, low-molecular-weight dyes (24).

Chemical Treatment Methods

- **Chemical Oxidation:** Chlorination and ozonation methods can cleave the dye. Ozone is more effective but has a short half-life and is more expensive. Chlorination may lead to the formation of toxic chlorinated organic compounds (25).
- **Advanced Oxidation Processes (AOPs):** Methods like Fenton's reagent (Fe^{2+}/H_2O_2), photocatalysis (e.g., TiO_2/UV), and sonolysis produce potent oxidizing agents like •OH that degrade dyes in a non-selective way. AOPs can be costly based on the chemicals used; however, the toxic nature of the intermediate (26).

Biological Treatment Methods

- **Aerobic and Anaerobic Microbial Degradation:** In this method, microbes degrade dyes as a source of carbon. This method is eco-friendly but it is a slow process because it is difficult to maintain conditions such as pH and temperature, and the rate of degradation depends on the dye. Most dyes are xeno-biotic and can only undergo partial degradation (27).

The limitations inherent in using these traditional approaches collectively, such as high costs, low efficiencies of specific dyes, secondary pollution, and complexity, bring up a technology gap. This gap propels the quest for the next generation of materials, which need to have high capacities, kinetics, selectivities, reactivities, and multifunctionalities.

GRAPHENE-BASED NANOMATERIALS

GBNs represent a family of materials derived from or related to the graphene sheet, each with distinct properties tailored for environmental applications (28).

Structure and Physicochemical Properties of Graphene

Fundamentally, graphene consists of a layer of carbon atoms arranged in a densely packed 2D hexagonal lattice structure with a bond length between carbon atoms of 0.142 nm. This sp^2 hybridized structure consists of a cloud of 'delocalized π electrons above and below the plane,' which explains its exceptional properties(29). For environmental remediation applications, some key properties are:

- **Ultra-High Specific Surface Area:** The theoretical value is as high as 2630 m^2/g, offering a tremendous opportunity for absorbing pollutants, far greater than that of activated carbon, which has a surface area of only 1000 m^2/g
- **Extraordinary Mechanical Strength and Flexibility:** Graphene has a high Young's modulus at ~1 TPa and high intrinsic flexibility, which makes it possible to fabricate strong and robust freestanding 3D architectures such as aerogels and sponges (30).
- **Superior Thermal and Electrical Conductivity:** These characteristics are of prime importance when GBNs are used as supports in photocatalytic/electrochemical treatment systems, where charge transfer is enabled, thereby avoiding electron-hole recombination (31).
- **Rich Surface Chemistry (particularly of GO):** GO's oxygen functionalities not only improve hydrophilicity and dispersibility in aqueous media but also offer the possibility of further functionalization and compositing with other polymers, metals, and metal oxides (32).

Types of Graphene-Based Nanomaterials

- **Pristine Graphene:** Pristine Graphene refers to perfect, defect-free monolayer material. In Pristine Graphene, despite the highest possible conductivity and theoretical surface area, there are very high intermolecular forces driving irreversible agglomeration in liquid medium, which limits its practical applications in water treatment processes (33).
- **Graphene Oxide (GO):** When graphite is oxidized strongly (for example, by the Hummers method), the resulting material is rich in epoxy, hydroxyl, carbonyl, and carboxyl functionalities. GO is consequently highly hydrophilic, more easily dispersed in solution, and has ample opportunity for electrostatic interactions and hydrogen bonding with dye molecules. Unfortunately, the disordered nature of the resulting carbon reduces its conductivity (34).
- **Reduced Graphene Oxide (rGO):** The chemical, thermal, or photocatalytic reduction of GO leads to the formation of reduced GO. Ideally, this is a compromise between the original graphene and GO. The reduction treatment removes a vast amount of the oxygen groups, which partially restores the conjugated sp^2 hybridization along with the conductivity, thus improving the π-π interactions with the aromatic dyes. The reduced hydrophilicity is still readily soluble and more processable compared to graphene itself (35).
- **Graphene Quantum Dots (GQDs):** These are graphene fragments that are nanometers in size, usually less than 20 nm, and show edge effects and quantum confinement. They have a high edge-to-area ratio with oxygenated groups, are water soluble, and are very fluorescent. Because GQDs can produce reactive oxygen species when exposed to light, they are being investigated further for sensing and photocatalytic degradation (36).

SYNTHESIS AND FUNCTIONALIZATION OF GRAPHENE-BASED NANOMATERIALS

The dye removal ability of GBNs is intrinsically related to their synthesis route and subsequent modification.

Synthesis Techniques

Top-Down Approaches:

- **Mechanical Exfoliation (Scotch Tape Method):** The first method produces good quality defect-free graphene flakes. However, the method is low throughput or labor-intensive and is not appropriate for large-scale production required for water treatment technologies (37).
- **Chemical Oxidation-Reduction:** This is the most common method for large-scale production. Modified Hummers' Method employs strong oxidizing agents such as $KMnO_4$, H_2SO_4, and $NaNO_3$ to produce GO. A subsequent reduction with hydrazine, sodium borohydride, or ascorbic acid results in rGO (38).
- **Electrochemical Exfoliation:** This is another promising eco-friendly process in which the electrode made of graphite is subjected to electrochemical potential in an electrolyte solution, thereby ensuring both intercalation and exfoliation into graphene and/or GO layers. This process is fast, scalable, and controllable in oxidation levels (39).

Bottom-Up:

- **Chemical Vapor Deposition (CVD):** It involves hydrocarbon gas (e.g., CH4) dissociating on a metal substrate (Cu or Ni) at high temperatures to prepare large-area, high-quality graphene films. Although it is used for electronics, it is not very significant in adsorbent preparation due to its cost (40).

Green Synthesis Approaches: Research emphasizes the use of environmentally friendly materials to address sustainability

- **Green Reduction of GO:** To produce rGO without using harmful reagents like hydrazine, plant extracts (such as aloe vera or green tea), vitamins (such as vitamin C), sugars (such as glucose), or microorganisms act as stabilizing and reducing agents (41).
- **Direct Biosynthesis:** Although it can be difficult to regulate the quantity and quality of layers, graphene-like materials can be produced by pyrolyzing or hydrothermally carbonizing biomass precursors (such as sugarcane bagasse or chitosan) (42).

Functionalization and Composite Formation

To improve the performance, prevent restacking, and add more functionality, GBNs are often modified.

- **Chemical/Surface Functionalization:** Functional groups of specific compounds covalently bound to the surface play a significant role in the modification of the surface chemistry. Amination (-NH2 groups) of the surface for increasing the positive charge for adsorbing anionic dyes or sulfonation of the surface (-SO3H).
- **Polymer-Graphene Composites:** The use of GBNs and polymers helps in the combination of the adsorbent's high specific surface area with the mechanical strength and flexibility of the polymer.
- **Chitosan-GO/rGO:** Chitosan is a biodegradable material that has hydroxyl (-OH) and amino (-NH2) functional groups. This biopolymer composite with GO has been found to improve the absorption capacity of the material. The composite has been found to improve the stability of the material in acidic media, in which the biopolymer is soluble (43).
- **Polyaniline (PANI)-Graphene:** PANI, which is a conductive polymer, has been used in composite form, where graphene conducts electrons for better electrochemical removal/photocatalysis (44).
- **Metal/Metal Oxide–Graphene Composites:** Decorating GBNs with nanoparticles creates multifunctional hybrids.
    - **Graphene functions** as an electron acceptor in photocatalytic composites (such as TiO_2/GO and ZnO/rGO), which prevents charge carrier recombination in the semiconductor and promotes photocatalytic dye degradation in the presence of UV and visible light (45).
    - **Magnetic Composites (e.g., Fe_3O_4/GO):** By incorporating magnetic nanoparticles (MNPs) such as magnetite (Fe_3O_4) or maghemite (γ-Fe_2O_3), these composites solve a crucial solid-liquid separation problem in real-world applications by enabling the quick separation of spent adsorbent from treated water using an external magnet (46).

• **Terrestrial and Hybrid Composites:** These cutting-edge systems integrate adsorption, photocatalysis, and magnetic separation into a single, recyclable material by combining several components, such as TiO_2-Fe_3O_4-rGO (47).

MECHANISMS OF DYE REMOVAL

The enhanced efficiency of GBN materials in dye sequestration can be attributed to a complex interplay among various physico-chemical processes, which may be either interacting or non-interacting. This effect may be dominated by either the physico-chemical nature of GBN materials or the solute dye structure and aqueous medium conditions (48).

Adsorption

- **π–π Stacking/Electron Donor-Acceptor Interactions:** Often times, this is usually the dominating mechanism when considering Aromatic Dyes on Graphene surfaces. The π-electron cloud surface on graphene interacts directly with the aromatic moieties of the dye molecule. In the case of graphene oxide (GO), its sp^2 domains usually function as an electron acceptor, while the dye molecules, which have an electron-rich ring system (e.g., Methylene Blue), function as an electron donor, enabling enhanced non-covalent adsorption mechanisms (49). When considering rGO surfaces, this is enhanced due to its more relaxed conjugated structure.
- **Electrostatic Attraction/Repulsion:** For ionic dyes, this is very important and pH-dependent. The point of zero charge (pHpzc), which measures the surface charge of GBNs, varies: GO's ionizable carboxyl groups make it negatively charged over a broad pH range, whereas rGO and functionalized graphenes can have a charge that can be adjusted. The surface is positively charged and protonated at pH < pHpzc, which

promotes the adsorption of anionic dyes (like Congo Red). The surface becomes deprotonated and negative at pH > pHpzc, which draws cationic dyes like Methylene Blue (50). If the charges are similar, electrostatic repulsion may prevent adsorption.

- **Hydrogen Bonding:** Compatible groups (-NH_2, -OH, -N=N-, -SO_3H) on dye molecules can form hydrogen bonds with the oxygen-containing functional groups (-OH, -COOH, C=O) on GO and functionalized GBNs. The adsorption of dyes such as Rhodamine B and Methyl Orange onto GO is greatly aided by this mechanism (51).
- **Van der Waals forces and hydrophobic interactions:** These weak, non-specific forces help non-ionic or hydrophobic dye molecules (like some disperse dyes) adhere to the less-oxidized, hydrophobic areas of graphene sheets or rGO (52).

Photocatalytic Degradation

Advanced oxidation is facilitated when GBNs are composited with semiconductor nanoparticles (such as TiO_2, ZnO, and CdS). The semiconductor produces electron-hole pairs when exposed to light. Photogenerated electrons are efficiently moved away from the semiconductor surface by the graphene component, which serves as a superb electron acceptor and transporter. This significantly lowers charge carrier recombination, which results in a longer hole (h^+) lifespan and a higher production of reactive oxygen species (ROS), such as superoxide anions ($\bullet O_2^-$) and hydroxyl radicals ($\bullet OH$) (53). Adsorbed dye molecules are subsequently non-selectively mineralized by these potent oxidants into CO_2, H_2O, and inorganic ions.

Synergistic Mechanisms in Hybrid Systems

Hybrid composites utilize synergistic "adsorb & degrade" or "concentrate & convert" mechanisms. For example, for the magnetic GO-Fe_3O_4-TiO

1. The GO sheets offer a high surface area for adsorbing dye molecules near the active sites primarily through π-π stacking and electrostatic interactions.
2. The anchored TiO_2 nanoparticles photocatalytically degrade the concentrated dyes
3. The Fe_3O_4 nanoparticles made it possible to separate the spent composite through magnetic separation.

This synergy achieves removal efficiencies that far exceed the cumulative results of the separate parts' performance levels (54).

FACTORS INFLUENCING DYE REMOVAL EFFICIENCY

In the optimisation of the dye removal process, operational parameters have to be controlled, as they significantly impact the mechanisms of interaction.

Solution pH

The most important parameter, as it controls the degree of ionization of the dye molecules and the surface charge of the GBN adsorbent. For instance, the adsorption capacity of the cationic dye, Methylene Blue, by GO is found to increase with an increase in pH, as the increased pH causes greater deprotonation of the carboxyl groups of GO, thus increasing the electrostatic attraction. The reverse is true in the case of the anionic dye, Congo Red (55). pH influences the stability of the GBN suspensions.

- **Contact Time and Adsorption Kinetics:** Efficiency tends to rise rapidly at first due to the availability of vacant adsorption sites, and then it slows down and reaches equilibrium. Fast adsorption kinetics are one of the characteristics of GBNs, which attain equilibrium in just a few minutes or hours, thus rendering it quite effective and advantageous from different points of view (56).
- **Initial Dye Concentration:** This is the driving force for mass transfer. Adsorption capacity increases with an increase in initial concentration except when the concentrations are quite high, in which case the amount of adsorbent becomes the driving factor (57).
- **Adsorbent Dosage:** An increase in adsorbent dosage means there are more available sites for adsorption; consequently, the rate of removal typically increases. However, there is a tendency for q_e to decrease due to agglomerates or sites being either unabsorbed or not saturated (58).
- **Temperature:** It affects both the kinetic and thermodynamic properties of the process. In general, higher temperatures increase the rate of diffusion. If the adsorption capacity increases with temperature, it is an endothermic process; if it decreases with temperature, then it is said to be exothermic, which implies a consideration for the amount of energy required during operation (59).
- **Ionic Strength and Co-Existing Ions:** Real wastewater contains a variety of salts and ions. High ionic strength compresses the electrical double layer, which could reduce the extent of electrostatic interactions. For example, competing cations such as Na^+ and Ca^{2+} can occupy negatively charged sites on GO, reducing the uptake of cationic dyes, and vice versa for anions and anionic dyes (60).

ADSORPTION KINETICS, ISOTHERMS, AND THERMODYNAMICS

It is, therefore, obvious that mathematical modeling of the experimental data is essential to understand the adsorption process for prediction and design of treatment systems.

Kinetic Models represent the rate of dye uptake and rate-controlling steps.

- **Pseudo-First-Order Model:** Assumes the rate of occupation of adsorption sites is proportional to the number of unoccupied sites. Mostly failed to fit the entire range of data for GBNs (61).
- **PSO Model:** It assumes that the rate of adsorption is proportional to the square of the number of unoccupied sites. It has been recently found to best describe the kinetics of dye adsorption onto many GBNs, which suggests that chemisorption (for instance, electrostatic attraction, hydrogen bonding) may be the rate-limiting step (62).
- **Intraparticle Diffusion Model (Weber-Morris):** Used to identify if pore diffusion is a controlling mechanism. A multi-linear plot often suggests adsorption occurs in stages: film diffusion

(external surface adsorption), followed by gradual intra-particle diffusion, and finally equilibrium (63).

- **Adsorption Isotherm** Models describe the equilibrium distribution of dye between liquid and solid phases at constant temperature.
- **Langmuir Isotherm:** This model is particularly suited for monolayer adsorption on a homogenous surface with identical non-interacting sites. A high fit to this Langmuir model (characterized by a separation factor RL where 0< RL <1) suggests that there are uniform finite sites on the GBNs, which is common in chemisorption (64).
- **Freundlich Isotherm:** This is a model for adsorption on heterogeneous surfaces. The Freundlich Isotherm suggests that the surface energy distribution is not uniform, which might be a feature for functionally modified GBNs (65).
- **Sips or Langmuir-Freundlich Isotherm:** A hybrid isotherm that has the advantage of combining both isotherms, especially when the system shows characteristics of both monolayer and heterogeneous isotherms at different concentration ranges (66).

The Thermodynamic Parameters give us an indication of the change in the processes of adsorption in terms of energy change and the spontaneity of the adsorption process itself, as resulting from experiments done at

- **Glossary Term:** Gibbs Law of Free Energy Change - ΔG°. ΔG° is negative for a spontaneous reaction. ΔG° becomes more and more negative as the temperatures increase for endothermic reactions (67).
- **Enthalpy Change (ΔH°):** If the ΔH° is positive, it indicates endothermic adsorption. In endothermic adsorption, capacity increases with temperature. This type of adsorption is associated with chemisorption. A negative ΔH° (68).
- **Entropy Change (ΔS°):** A positive value of ΔS° is a criterion showing an increase in randomness at the solid solution interface during adsorption; usually, a consequence of the desorption of a layer of water molecules from the adsorbent's surface by the adsorbate/dye molecules (69).

REGENERATION, REUSABILITY, AND STABILITY

For economic viability and environmental concerns of used adsorbents, reusability is important.

- **Desorption and Regeneration Methods:** The desorption method depends on the main mechanisms of adsorption.
- **Solvent Elution:** Organic solvents (ethanol, methanol) or acidic/alkaline solutions (HCl, NaOH) can be employed. For cationic dyes adsorbed electrostatically by negatively charged GO, an acidic eluent of pH ~2-3 can be used as the electrostatic attraction between the oppositely charged species is repelled in an acidic medium, where the protons will have the same charge as the dye on the negatively charged GO.
- **Photocatalytic Self-Regeneration:** For photocatalytic adsorbents (e.g., TiO2/rGO), self-regeneration of the adsorbent is achieved by exposing the dye adsorbed photocatalytic composite to light, thus destroying the adsorbed dye and renewing the adsorbent surface.
- **Reusability Performance:** An important criterion for application. In most research, performance is evaluated over a period of 4-6 cycles. Good GBN composites possess the ability to retain >70-80% of the original capacity. Magnetic composites have good prospects since their separation mechanism prevents any physical loss from occurring (70).
- **Structural and Chemical Stability:** Physical disintegration or chemical degradation (such as the loss of functional groups) can result from repeated adsorption-desorption cycles. After cycling, characterization using FTIR, XRD, and SEM is essential to verifying the material's resilience. Longevity can be greatly increased by stable covalent functionalization or cross-linking in polymer composites (71).

ENVIRONMENTAL AND TOXICOLOGICAL ASPECTS

An important but sometimes overlooked aspect of GBNs' use is their potential release into ecosystems during the course of their lifespan, which calls for a precautionary evaluation of their environmental safety.

- **Environmental Fate and Behavior:** GBNs' physicochemical characteristics and the surrounding environment determine how they disperse, aggregate, move, and ultimately end up in aquatic systems. Because GO is hydrophilic, it may create stable colloidal solutions, which may enable long-distance transportation. rGO, on the other hand, has a tendency to settle and aggregate. These behaviors can be significantly changed by the presence of natural organic matter (NOM), ionic strength, and pH, which can either stabilize or destabilize the dispersions (72).
- **Ecotoxicological Concerns:** GBN toxicity to aquatic life depends on both dosage and property.
- **Physical Damage:** Cells, such as bacteria and algae, may sustain physical membrane damage as a result of sharp sheet edges (73).
- **Oxidative Stress:** GBNs, particularly GO, have the ability to cause organisms to produce reactive oxygen species (ROS), which can result in DNA damage, protein oxidation, and lipid peroxidation (74).
- **Trophic Transfer:** There is evidence that GBNs may be absorbed by algae, which are primary producers, and then passed on to fish and zooplankton, which are higher trophic levels. This might lead to bioaccumulation (75).
- **Human Health Implications:** Occupational exposure during manufacturing or handling is a primary concern. Inhalation of graphene particles may cause pulmonary inflammation and fibrosis, analogous to other nanomaterials. Dermal exposure and potential ingestion routes also require thorough investigation (76). Functionalization can mitigate toxicity; for instance, PEGylation (attachment of polyethylene glycol) is known to improve biocompatibility.

COMPARATIVE ASSESSMENT WITH OTHER ADSORBENTS

Table 1

A Critical Comparison Situates GBNs within the Broader Landscape of Water Treatment Materials.

Adsorbent Category	Examples	Key Advantages	Key Limitations vs. GBNs
Conventional Carbon	Activated Carbon (AC), Biochar	Low cost, widely available, high porosity.	Lower specific surface area than graphene; slower kinetics; difficult and costly regeneration; non-tunable surface chemistry.
Other Carbon Nanomaterials	Carbon Nanotubes (CNTs)	High surface area, good mechanical strength.	Generally more expensive to produce; prone to bundling; lower adsorption capacities reported for some dyes; greater environmental toxicity concerns [82].
Clay & Minerals	Bentonite, Kaolinite, Zeolites	Abundant, very low cost, ion-exchange capacity.	Much lower adsorption capacity and slower kinetics; limited stability in wide pH ranges; less versatile surface for functionalization.
Agricultural / Industrial Waste	Rice husk, Fly ash, Sawdust	Extremely low cost, promotes waste valorization.	Highly variable and inconsistent composition; low to moderate capacities; often require extensive pretreatment; may leach color/organics.
Metal-Organic Frameworks (MOFs)	ZIF-8, MIL-101	Extremely high porosity & surface area; highly tunable.	Poor stability in water for many types; high synthesis cost; complex regeneration; potential metal leaching.
Graphene-Based Nanomaterials	GO, rGO, Composites	Ultra-high surface area; excellent & tunable surface chemistry; fast kinetics; high capacity; versatile for composites (magnetic, photocatalytic).	Current high production cost at scale; potential toxicity; aggregation in aqueous media without functionalization.

Conclusion of Comparison

GBNs are repeatedly shown to have quicker kinetics than AC and better or equivalent adsorption capabilities, frequently orders of magnitude greater than clays and biowaste. They are superior to most traditional alternatives due to their multifunctionality, which includes the capacity to be designed as adsorbents, catalysts, and readily separable materials. The primary obstacles are environmental and economic in nature rather than performance-related (77).

CHALLENGES, RESEARCH GAPS, AND FUTURE PERSPECTIVES

Although the development of GBNs has been highly successful at the laboratory scale, their application at the field scale is considered a major challenge.

Scalability and Economic Feasibility

Economically viable, scalable, and green synthesis of superior-quality GBNs at large scales is still in its infancy. Although the existing chemical approaches, namely Hummers', are dangerous, the 'green techniques' need to be optimized. Moreover, an environmentally friendly life cycle assessment is essential to assess the true cost of GBN-based systems in comparison with other existing approaches (78).

Performance in Real Wastewater Matrices

More than 95% of the literature utilizes synthetic solutions containing dyes. Real-world wastewaters are complex mixtures of different dyes, salt, acid/base, metal, surfactants, and organics. The research needs a shift towards assessing performance under such realistic conditions, wherein the presence of competing dyes, fouling, and pH can significantly lower the GBNs, as demonstrated elsewhere (79).

Sustainability and Lifecycle Management: A green application does not have a perfectly unsustainable lifecycle either. Future work needs to be done on:

- **Sustainable Feedstocks:** To produce graphene, we could use biomass carbon.
- **Design for Regeneration & End-of-Life:** Designing GBNs which are not only regenerable but ultimately biodegradable or recyclable without causing pollution to the environment itself (80).

Regulatory and Standardization Gaps: There are no standard protocols for evaluating the environmental fate, ecotoxicity, and stability of GBNs in water treatment operations. The regulations concerning the use of nanomaterials in the environment and their applications remain in their infancy stage.

Future Research Directions

1. Engineering Macroscopic Architectures: Breaking through the barrier of powders towards robust, reusable three-dimensional macrostructures such as graphene aerogels, hydrogels, sponges, and porous monoliths suitable for fixed-bed or column flow-through configurations, which are of greater technological interest compared to powders (81).

2. Computational-Guided Design: Molecular dynamics (MD) and density functional theory (DFT) simulations for predicting the interaction between dyes and GBNs at the atomic level, which would facilitate rational-based design with high selectivity towards certain dye molecules (82).

3. Multifunctional "Smart" Systems: Developing composites which unite the power of adsorption, in situ degradability (photo, electro,

4. Long-Term Environmental Impact Studies: Carrying out extensive, realistic studies to comprehend long-term exposure, fate, and chronic effects of GBNs released into the environment.

CONCLUSION

Graphene-based nanomaterials have unquestionably emerged as a game-changing and powerful class of materials to combat the chronic problem of dye pollution in wastewater. This review has integrated a massive volume of knowledge that the unprecedented performance exhibited by GBNs originates from an unbeatable synergy between its ultra-high surface area, tunable surface chemistry rich in π-electrons and functional groups, and an unprecedented ability to form synergistic composites with polymers, metals, and metal oxides. These characteristics allow highly efficient dye removal through a combination of mechanisms:

adsorption via π-π stacking, electrostatic forces, and hydrogen bonding, besides photocatalytic degradation in advanced composites.

The implications are significant, as GBNs promise the opportunity to deliver quicker, more powerful, and more flexible treatment systems than those afforded by conventional treatment options like activated carbon filtration or coagulation. Yet, the journey from promising nanomaterial to viable environmental technology is complex. As it currently is, the environmental technology landscape can be characterized by a striking disparity between outstanding laboratory results and ongoing challenges with green-scale-up processes, effectiveness and stability in challenging waste matrices, environmental risk assessment, and cost competitiveness.

This future perspective, then, has to be one of convergent research. Material scientists would need to collaborate with environmental engineers, toxicologists, and industry stakeholders. There is a need to shift the perspective from simply emphasizing high adsorption capacities for pure water to engineered material systems which are demonstrated under realistic conditions. This critical bridging, then, would ensure that graphene-based nanomaterials do not remain confined to very active research interest but can progress to become a cornerstone technology for delivering sustainable water management strategies, thereby making a positive contribution to safe global water resources and environmental security.

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